Thermal hysteresis in the normal-state magnetization of $La_{2-x}Sr_xCuO_4$

C. Panagopoulos,[1,2] M. Majoros,[2] and A. P. Petrović[1,2,*]
[1]*Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, United Kingdom*
[2]*Interdisciplinary Research Centre in Superconductivity, University of Cambridge, Cambridge CB3 0HE, United Kingdom*
(Received 27 August 2003; revised manuscript received 11 November 2003; published 14 April 2004)

We have performed magnetization measurements on $La_{2-x}Sr_xCuO_4$. A hysteresis in the temperature dependence of low field magnetization is observed well above the superconducting transition temperature. The behavior of the hysteresis resembles fundamental properties of the superconducting state and is discussed in terms of superconducting correlations.

DOI: 10.1103/PhysRevB.69.144508 PACS number(s): 74.72.Dn, 74.25.Ha, 74.25.Qt

Raising the superconducting transition temperature (T_c) has been a major scientific challenge since the discovery of superconductivity in Hg ($T_c = 4.2$ K). High-temperature superconductivity (HTS) in copper oxide superconductors with $T_c = 30$ K raised hopes that this class of oxides may be the key to materializing yet another revolution in science.[1] However, the highest T_c at ambient pressure achieved to date is 135 K.[2] On the other hand, recent magneto-thermal transport experiments on $La_{2-x}Sr_xCuO_4$ suggested onset temperatures of vortex excitations well above T_c, namely, at 130 K for a HTS family with maximum $T_c = 39$ K.[3,4] Similar results on other HTS indicate the generic behavior of this effect.[4]

Here, using straightforward magnetic measurements on $La_{2-x}Sr_xCuO_4$ we find evidence for the presence of hysteresis in the temperature dependence of low field magnetization up to 290 K. The temperature, field, and crystallographic (ab plane and c axis) dependences of the onset and strength of the hysteresis resemble fundamental properties of the superconducting state. Our results suggest a connection between the observed hysteresis and superconducting correlations.

The materials studied were $La_{2-x}Sr_xCuO_4$ with $x = 0.03$, 0.06, 0.07, 0.08, 0.10, 0.11, 0.125, 0.135, 0.17, 0.20, 0.22, and 0.24 (among these the $x = 0.03$ samples were both polycrystals and single crystals, the $x = 0.11$ only single crystals, and all others polycrystalline). The $La_{2-x}Sr_xCuO_4$ powders and single crystals were prepared using solid-state reaction procedures and the traveling solvent floating-zone technique, respectively. No traces of impurities were detected from spectroscopic and chemical analyses. Scanning electron microscopy showed that the grains in the polycrystalline samples were approximately spherical with average grain diameter $\sim 7\ \mu$m. The dimensions of the $x = 0.03$ crystal were $5 \times 2.5 \times 1.5\ mm^3$ whereas the $x = 0.11$ was a cylinder with 5 mm diameter and 2 mm thickness. Magnetization (M) measurements were performed as a function of temperature (T) and field (H) using a quantum design (MPMS-XL) superconducting quantum interference device (SQUID) at a 3-cm scan length. Results were also checked using 6-cm scan length. Where sensitivity was allowed, some of the results were confirmed using an earlier model of this instrument (MPMS). In a typical M-T run we first zero field the magnet (this procedure leaves a trapped field $< \pm 1.5$ G) and the sample cooled to 5 K. The desired field was applied $H > H_p$, where H_p is the applied magnetic field at which flux penetrates for the first time upon increasing the magnetic field, and the sample warmed slowly to room temperature and to 320 K for $La_{1.9}Sr_{0.1}CuO_4$ [zero-field cooled (ZFC)]. The sample was then cooled back to 5 K in the presence of the applied field [field cooled (FC)] and at the same temperature rate. The temperature sweeps were performed at 1 K/min to assure thermal equilibrium. The results were confirmed under different heating and cooling rates (0.5, 2, and 4 K/min). The background due to the sample holder was subtracted for each run. Many of these early results were confirmed by later measurements, reported in this paper, where no extra material was added to hold the sample, and also no discontinuity in the sample holder was present apart from the sample. It is the discontinuity which is responsible for the SQUID signal and, therefore, in the experimental configuration with no discontinuity in the sample holder the measured signal reflects purely the sample's magnetization. To ensure the intrinsic nature of the findings measurements were performed on several samples prepared in different laboratories.

In Fig. 1 we present typical M-T data for superconducting $La_{2-x}Sr_xCuO_4$ (LSCO) samples with $x = 0.06$, 0.07, 0.10, 0.135, 0.22, and 0.24 measured at $H = 100$ G. The normal state susceptibility is seen to follow the known doping dependence.[5] However, for $0.06 \leq x < 0.24$ a small hysteresis develops at temperatures T_s, indicated by arrows in the figure, several times the bulk superconducting transition temperature T_c (values of T_c are shown in Fig. 9), and at room temperature for $x = 0.10$. We also notice a small but smooth increase in the hysteresis width as T_c is approached, indicating a connection to the mixed state (see, e.g., plots for $x = 0.07$, 0.10, and 0.22). Note that for clarity we only show data from just above T_s. Data at $T \gg T_s$ displayed no hysteresis at the level shown for $x = 0.24$ in Fig. 1. Although our sample quality tests indicate all samples measured are pure and of the highest quality we are unable to confirm that the results shown in Fig. 1 unambiguously reflect quantitatively an ideal impurity/disorder free HTS. However, based on (i) our sample characterization procedures, (ii) the agreement between results obtained on samples prepared by different groups and in different laboratories, and (iii) the data sets discussed below, we are confident that the overall trend re-

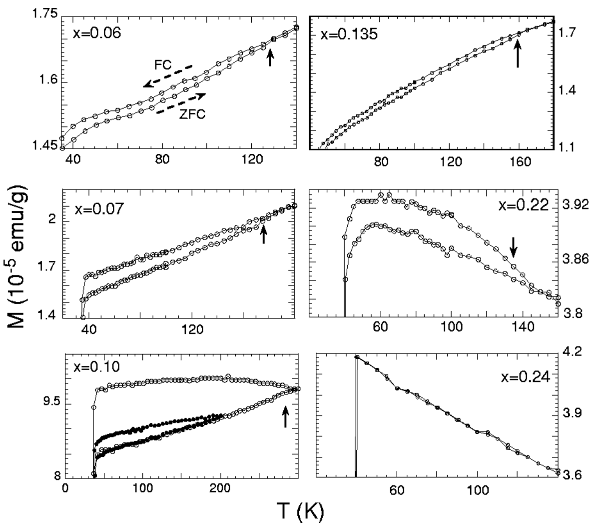

FIG. 1. Plots of zero-field-cooled (ZFC) and field-cooled (FC) magnetization in polycrystalline $La_{2-x}Sr_xCuO_4$ at different values of x measured at $H = 100$ G. The panel for $x = 0.10$ includes an M-T cycle (closed circles) only up to 200 K (see text for details). Solid arrows indicate the onset temperature of the hysteresis T_s. The lower curves in the hystereses are always the ZFC sweep.

ported here is at least qualitatively representative of this high-T_c cuprate.

To examine further the behavior of the hysteresis and T_s we have performed measurements on single crystals ($x = 0.11$). In Fig. 2(a) we show data for $H(\| ab)$ $= 5$ G ($< H_p \cong 20$ G, measured for $H \| ab$ at $T = 5$ K). Here we expect no vortices to penetrate the sample at $T = 5$ K, the

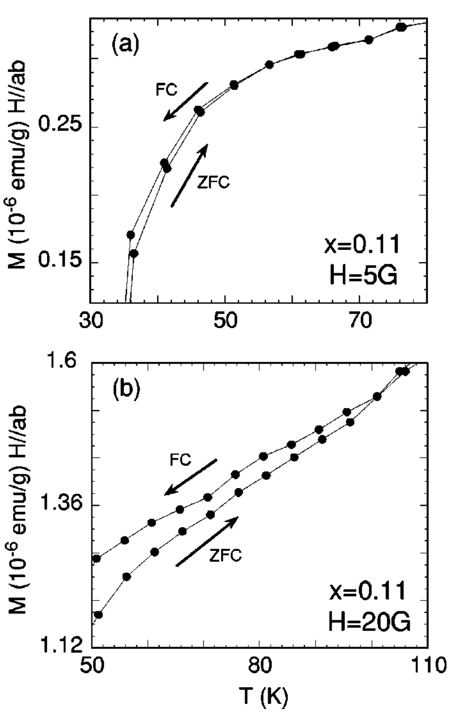

FIG. 2. Magnetization as a function of temperature at (a) $H = 5$ G and (b) $H = 20$ G for a $La_{2-x}Sr_xCuO_4$ ($x = 0.11$) single crystal with $H \| ab$.

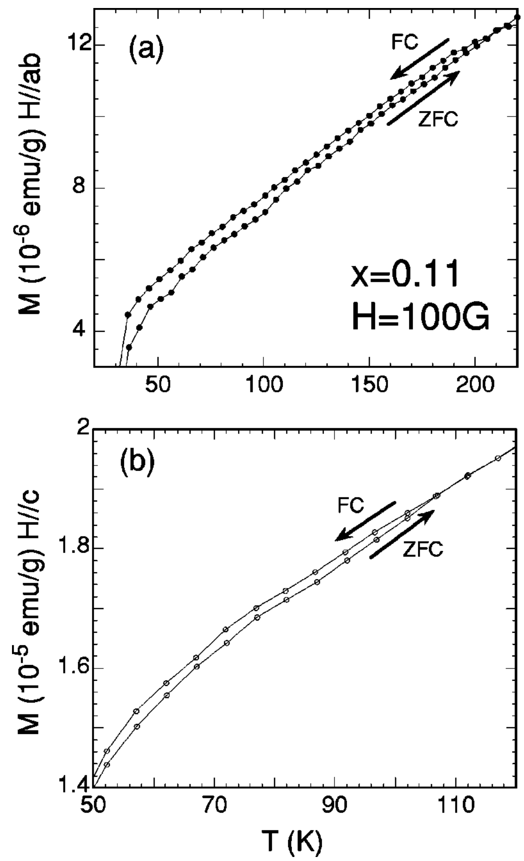

FIG. 3. Magnetization as a function of temperature at $H = 100$ G for a $La_{2-x}Sr_xCuO_4$ ($x = 0.11$) single crystal for (a) $H \| ab$ and (b) $H \| c$.

temperature the ZFC measurement commences. We do, however, anticipate flux penetration as the temperature is raised and H exceeds $H_p(T)$, in which case vortices will be only weakly trapped. Figure 2(a) indicates the presence of a tiny thermal hysteresis above T_c with $T_s \cong 50$ K and a hysteresis width $\Delta M/H \cong 4 \times 10^{-10}$ emu/G at $T = 40$ K. In Fig. 2(b) we present results for $H(\| ab) = 20$ G which is nearer H_p. In this case $T_s \cong 90$ K, and e.g., $\Delta M/H \cong 1 \times 10^{-8}$ and 2×10^{-9} emu/G at $T = 40$ and 60 K, respectively.

Increasing the applied field to $H(\| ab) = 100$ G we obtain $T_s \cong 200$ K and $\Delta M/H \cong 1.3 \times 10^{-8}$ and 4×10^{-9} emu/G at $T = 40$ and 60 K, respectively [Fig. 3(a)]. At significantly higher applied fields, $\Delta M/H$ and T_s drop again (Fig. 4). The inset in the lower panel of Fig. 4(b) shows a similar field dependence for ΔM and T_s for polycrystalline LSCO ($x = 0.10$).

Measurements for $H \| c$ at $H = 100$ G ($> H_p \cong 35$ G, measured for $H \| c$ at $T = 5$ K) indicate an anisotropic behavior of the hysteresis [Fig. 3(b)]. Figures 3 and 4 show that for $H \| ab$, ΔM is wider and T_s is higher and is suppressed more slowly by field. The above tests indicate the presence of an anisotropy in ΔM and T_s (Fig. 3) and that a well-defined hysteresis and high values of T_s are observed only when H is applied well above H_p and at $T \ll T_c$ (Figs. 2 and 3). On the other hand, $H \gg H_p$ leads to a gradual suppression of T_s and $\Delta M/H$ (Fig. 4).

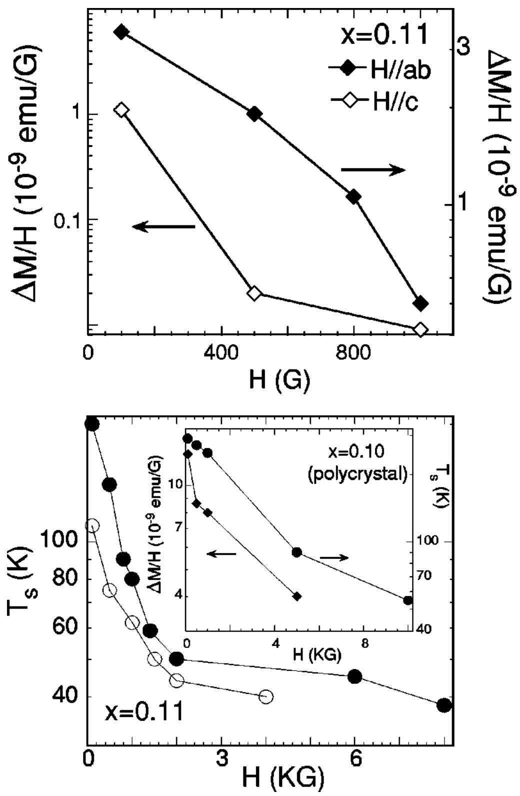

FIG. 4. The upper panel shows the field dependence of the hysteresis width $\Delta M/H$ for the single crystal for the two field orientations determined at $T=60$ K. In the lower panel is a plot of T_s again for the two field orientations. Closed and open symbols represent $H\|ab$ and $H\|c$, respectively. The inset in the lower panel shows T_s and $\Delta M/H$ (determined at $T=70$ K) for a polycrystalline sample with $x=0.10$.

In Fig. 5 we show the hysteresis for $H\|ab$ but nearer T_c, indicating an increase in the width with approaching the mixed state, in agreement with the polycrystalline samples in Fig. 1. Similar behavior is reported below for other materials [Fig. 8(b)]. The smooth evolution of the hysteresis may be taken as suggestive of a common cause for the hysteresis

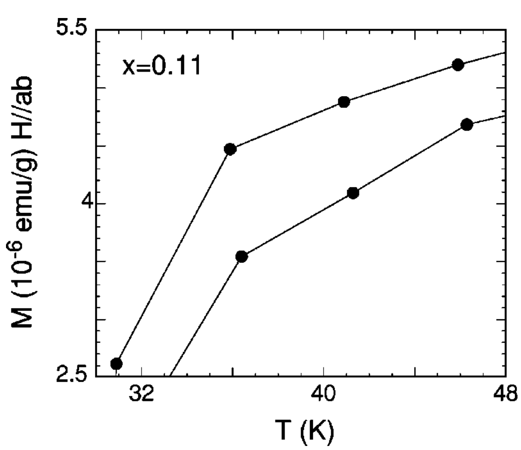

FIG. 5. A zoom-in plot near T_c of the magnetization as a function of temperature at $H=100$ G for a $La_{2-x}Sr_xCuO_4$ ($x=0.11$) single crystal for $H\|ab$.

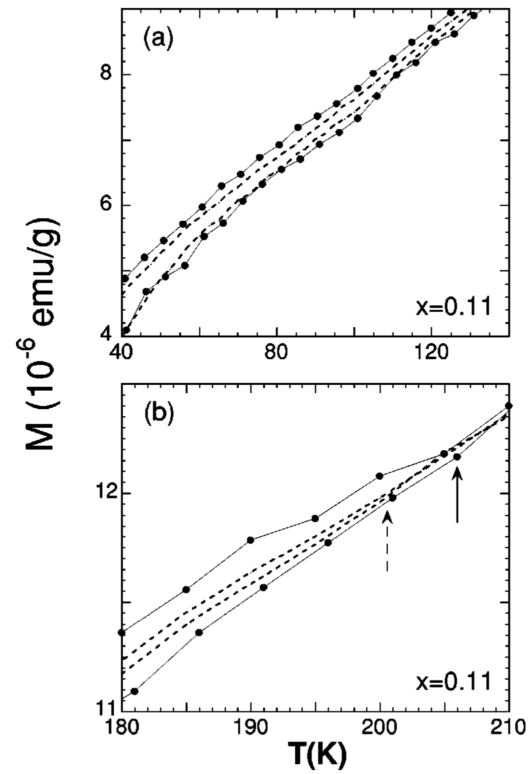

FIG. 6. Magnetization as a function of temperature at $H=100$ G for a $La_{2-x}Sr_xCuO_4$ ($x=0.11$) single crystal with $H\|ab$ (circles) [the same data as shown in Fig. 3(a)] and with H approximately 3° off the ab plane of the crystal (broken line). The data for the off-plane measurements (broken line) have been shifted downwards by 5×10^{-7} emu/g for clarity. Panel (a) shows data nearer T_c, whereas (b) near T_s. Note the small but distinct decrease in ΔM and the accompanied drop in T_s (b) (indicated by arrows).

below and above T_c, bearing in mind that the cause for a hysteresis below T_c is the presence of pinned superconducting vortices.

The sensitivity of T_s and ΔM on the crystallographic orientation can be seen also in Fig. 6, where we compare data for $H\|ab$ [same data as shown in Fig. 3(a)] with results obtained by remeasuring the sample with H approximately 3° off the ab plane of the crystal. The results obtained with the applied field ~3° off plane are shown as broken lines. Figure 6(a) presents the data near T_c, whereas Fig. 6(b) near T_s. We observe a small but distinct decrease in ΔM and as shown in Fig. 6(b) this is accompanied by a drop in T_s. (Note that the data for the off-plane measurements have been shifted downward by 5×10^{-7} emu/g for clarity.) These results indicate the gradual decrease of the hysteresis as H deviates from the CuO_2 planes. Notably, this trend is similar to that of the irreversibility field H_{irr} in the mixed state.[6]

Although the thermal hystereses we observe survive well above T_c, it is useful to mention a few similarities with properties occurring in the mixed state of these materials. The field orientation ($H\|ab$) for which T_s is higher and stronger against H is the orientation for which vortex pinning below T_c is strongest due to intrinsic pinning.[5] Also, $T_s(H)$ resembles the field dependence of the irreversibility tempera-

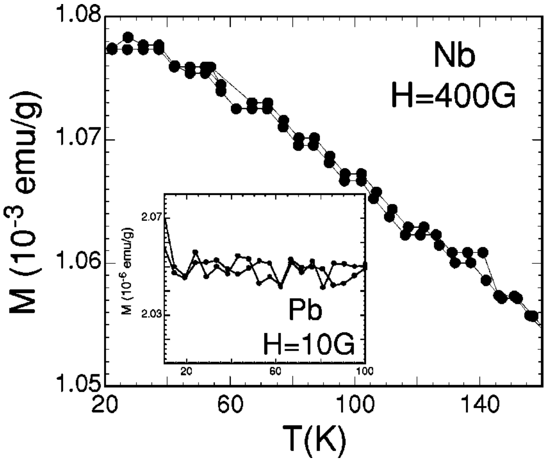

FIG. 7. Magnetization as a function of temperature for bulk Nb and Pb (inset). Measurements were performed at the fields indicated in the figure.

ture T_{irr}.[5,6] The orientation dependence of the width and the onset of the thermal hysteresis are also in broad agreement with the width and the onset of the magnetic hysteresis in M-H measurements we performed in the mixed state. For example, for the $x=0.11$ single crystals and at $T=22$ K we obtain $H_{irr}=4.5$ and 0.3 T for $H\|ab$ and $H\|c$, respectively. These trends agree with the HTS literature, where the maximum in the critical current density, and irreversibility field and temperature when $H\|ab$, is attributed to intrinsic pinning.[5–7] Of course the concept of pinned vortices surviving to very high temperatures, and in particular at $T>T_c$, is by no means conventional. However, it is worth mentioning that crystals of $YBa_2Cu_3O_{7-\delta}$ with $T_c=90.4$ K exhibit intrinsic pinning even at $T=89.8$ K, indicating the latter is not a phenomenon restricted to very low temperatures.[8]

The hystereses reported in this work are small. However, they are at least an order of magnitude larger than the level of the experimental noise. Furthermore, we would like to emphasize that we never failed to reproduce the results obtained, for any of the samples studied, once critical parameters, such as the heating/cooling rate, the magnitude of applied field, and the temperature the field was switched on for the ZFC run, were optimized.

It is unlikely our observations are due to extrinsic magnetism. For the single crystal, for example, the virgin curve in M-H measurements with $H\|c$, at, e.g., $T=50$ K, measured by cooling the sample to 5 K and then warming to 50 K in zero field, was linear in field, exhibited no residual magnetization and no detectable hysteresis was observed. (We note, however, that unlike other dopings the Sr=0.10 sample, which exhibits the largest hysteresis width and T_s, did display signatures for the possible presence of weak ferromagnetic behavior.) We have checked for possible surface, and bulk homogeneity across the sample, dependence of our data by cutting a smaller piece of the crystal and polishing it. The results for the two field orientations were in excellent agreement with the data for the uncut sample. Furthermore, as shown in Fig. 7, similar measurements on bulk Nb and Pb samples exhibited no hysteresis. (The temperature dependence and magnitude of M/H are in agreement with the literature.[9]) The M-T data shown here for Nb were taken for $H=400$ G ($H_p=30$ G) and for Pb at $H=10$ G. We have also checked for possible hysteresis using other fields. Again, we found no hysteresis for similar measurements for $H=50$, 100, 800, and 2500 G for Nb and $H=30$ G for Pb, confirming that there is no thermal hysteresis in the normal state magnetization of these two superconductors.

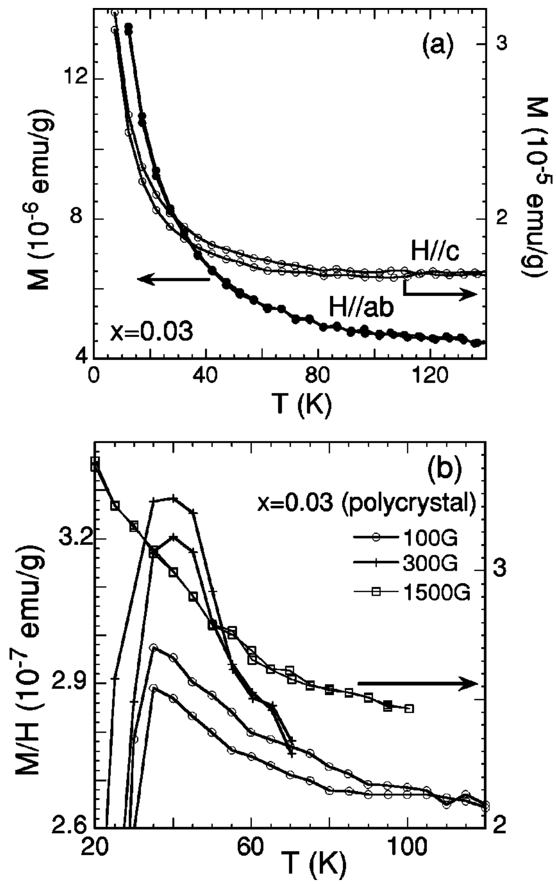

FIG. 8. (a) Plots of the anisotropic magnetization as a function of temperature for a $La_{2-x}Sr_xCuO_4$ ($x=0.03$) single crystal measured at $H=100$ G. (b) The susceptibility for an $x=0.03$ polycrystalline sample with filamentary superconductivity showing the gradual suppression of the hysteresis as superconductivity below T_c is suppressed at the same time. The lower curves in the hystereses are the ZFC sweeps.

We also note the agreement of our LSCO data with local magnetic imaging (scanning SQUID microscopy) on underdoped LSCO thin films ($T_c=18$ K), where the experimental resolution allowed the observation of local diamagnetic regions persisting up to 80 K.[10] Based on the experimental trends, the resemblance of the hysteresis in the normal state with that below T_c is interesting and the connection to superconductivity and the possible presence of persistent internal screening currents and perhaps even pinned vortices at these high temperatures appealing. It is therefore instructive to perform further tests.

Figure 8(a) shows M-T data taken for a nonsuperconducting LSCO ($x=0.03; T_c=0$) single crystal at 100 G for $H\|ab$ and $H\|c$. There is a hysteresis in M-T for $H\|c$, as expected

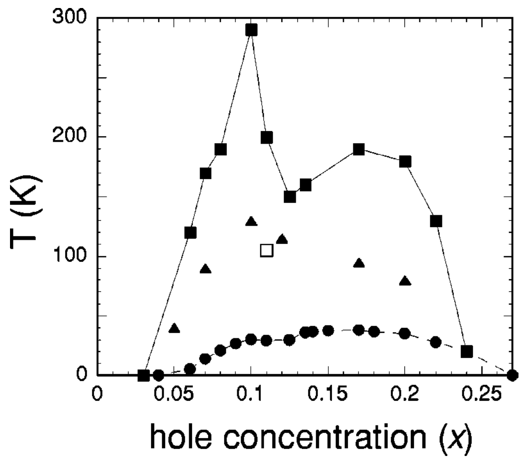

FIG. 9. The doping dependence of the superconducting transition temperature T_c (circles), the maximum onset of the hysteresis T_s (closed squares), and the onset temperature T_{Nernst} (triangles) of vortexlike excitations determined from Nernst effect studies with $H \| c$ (Refs. 3 and 4) for $La_{2-x}Sr_xCuO_4$. For $x=0.11$, T_s values shown as closed and open squares are for $H \| ab$ and $H \| c$, respectively. The error in the values of T_s, when $T_s > T_c > 0$, is ± 10 K.

from intrinsic weak ferromagnetism at this doping content and field orientation in LSCO.[11] However, there is no comparable hysteresis for $H \| ab$: the field orientation, the strength, and the magnitude of the hysteresis and T_s are maximum for the superconducting crystal ($x=0.11$) (Figs. 3 and 6). We notice, however, the presence of a very weak hysteresis for $H \| ab$ below 25 K, indicating the onset of the observed thermal hysteresis in the susceptibility is not abrupt at $x>0.05$ where bulk superconductivity sets in. To test the irrelevance of the hysteresis seen in the $x=0.03$ crystal for $H \| c$ to those seen in superconducting samples, we have performed similar measurements on polycrystalline LSCO again with $x=0.03$, but with excess oxygen incorporated in the lattice allowing presence of filamentary superconductivity. We find that when filamentary superconductivity is present a hysteresis is observed in M vs T below $T_s=110$ K [Fig. 8(b)]. Partial suppression of filamentary superconductivity using $H=300$ G yields a suppression in the hysteresis, which collapses fully at higher fields where superconductivity is also fully suppressed. These results indicate the presence of superconductivity is essential for the existence of a hysteresis above T_c.

In Fig. 9 we summarize the doping dependence of T_s for LSCO. Let us note that by considering the experimental noise, as well as the slight variation in the T_s values we obtained from measurements performed on different samples but for the same Sr concentration, the error in T_s is estimated to be ± 10 K. Comparison with the onset temperature T_{Nernst} (measured with $H \| c$) of the vortexlike signal[3,4] indicates correlation between the two energy scales reinforcing the superconducting nature of the hysteresis. We also note the excellent agreement between T_{Nernst} and T_s near $x=0.11$ ($H \| c$). Here we should also mention that although the agreement between the two techniques is very good, the two methods should be viewed at best as complementary, since the experimental configuration is different and they are sensitive to distinct energy scales with the Nernst effect requiring mobile rather than pinned vortices.

We would like to draw the attention on how well $T_s(x)$ tracks $T_c(x)$ with changes in the former being sharper. Furthermore, the higher the $T_s(x)$ the larger the maximum ΔM for a given sample (see, for example, Fig. 1) and the field required to suppress them. From Fig. 9 we identify another useful result. The suppression of superconductivity in the 1/8 region (plateau in T_c), believed to be related to the presence of stripes[12–14] is now seen more clearly in the doping dependence of T_s. We also notice that for $x>0.125$, T_s never recovers to high values. In fact, measurements of the absolute value of the superfluid density[15] $\rho_s(0)$, in the same system showed a dip at 1/8 with $\rho_s(0)$ never recovering to the extrapolated value at higher dopings, as expected by the Uemura relation $[\rho_s(0) \sim T_c]$,[16] just like T_s. These results add further credence to the relation of T_s to superconductivity.

Figures 1 and 9 show $T_s = T_c$ at $x=0.24$. We do not understand why $T_s = T_c$ at this specific doping; however, we speculate that at $x \geqslant 0.24$ either increased scattering has suppressed the hysteresis or it is an effect arising from drastic changes occurring at the Fermi surface of LSCO[17] and the rapid depletion of $\rho_s(0)$ at $x>0.24$.[18] Although there is no published report on Nernst effect studies for $x \geqslant 0.24$ our work has motivated the Princeton group[3,4] to perform Nernst measurements on single crystals. In agreement to our hysteresis work they find $T_{\mathrm{Nernst}}=0$ for $x>0.24$.[19]

It is unlikely internal short-range magnetism is responsible for T_s. Systematic studies have shown a spin glass regime, and associated high-temperature spin fluctuations, in LSCO and other HTS to develop gradually for $x<0.19$,[15] unlike the doping dependence of T_s. We can also rule out the possible effect of impurities and defects, which might have gone undetected. If these exist they must be present in all the materials we have investigated and satisfy the observed T, H, x, and crystallographic dependences, as well as the reported connection to superconductivity and correlation to other studies. We have also tested our results for age effects by measuring all samples more than once over a period of 12 months and found no change. Therefore, on the basis of the tests we have performed and the different sources, crystallinity, and preparation conditions of the series of samples investigated, we believe our results are not due to an extrinsic cause which has gone undetected. We are confident that the phase diagram shown in Fig. 9 is at least qualitatively representative of LSCO.

We appreciate that discussing these high temperature effects in terms of vortices is unorthodox. In particular, if superconducting fluctuations survive up to high temperatures we would expect the effect to be stronger for $H \| c$. This puzzle has driven us in performing the series of tests we report. It is due to these tests and the observed effects, which are so robust and closely linked to the creation of vortices at $T<T_c$, that we find it difficult to ignore the tendency of a possible connection between the hystereses, vortices, and superconductivity. Furthermore, magnetization is a bulk measurement with the measured signal reflecting the sample it-

self, and the data analysis straightforward and particularly sensitive and instructive to magnetism and superconductivity.

Of course, if there is superconductivity present at $T>T_c$ we may ask why we do not observe a diamagnetic signal in our uniform magnetic susceptibility. Again if we adopt a vortex scenario this, as well as the absence of zero or significantly reduced resistivity, may be understood in terms of the very small number of excess vortices relative to antivortices, embedded into the bulk paramagnetic background. For example, for $x=0.10$ where the effect is strongest, and for which a small drop in resistivity is actually seen at these high temperatures and sometimes attributed to precursor superconductivity,[20] we estimate only $\sim 10^4$ flux quanta at 100 K in 24 mm^3. Therefore, for a given $H>H_p$, only a very small number of pinned vortices may survive up to T_s, leading to a reduction in the magnitude of the paramagnetic signal (ZFC sweep). The FC curve, starting from $T>T_s$, reflects the fully paramagnetic response of the sample, i.e., in the absence of pinned vortices. To test this scenario we kept the field on after a FC run and warmed the sample to 300 K again. We obtained a curve identical to that of the previous FC run. Cooling back again with the field on tracked the FC curve once again. Further support comes from M-T cycles up to $T \ll T_s$. A typical set of data measured again at 100 G is shown for $x=0.10$ in Fig. 1. In this case the FC curve is lower than that obtained when the ZFC sweep crossed T_s (same panel in Fig. 1 but open circles). This difference may be understood if less field is trapped in the sample when we warm to $T \ll T_s$. Similar behavior is seen in experiments conducted on the same sample, as well as the single crystal ($x=0.11$), but in the superconducting state with M-T cycles up to $T<T_{\mathrm{irr}}$. This is a well-known behavior in HTS and points towards a similarity in the behavior of the hysteresis below and above T_c.

Based on the common effects we have observed among various materials, the dependence of the thermal hysteresis on superconductivity, the similarity between the crystallographic orientation dependence of ΔM and T_s and mixed state properties, and the resemblance between T_s and T_{Nernst}, we are inclined to believe we have observed an effect linked to some form of superconducting correlations. To account for the thermal hysteresis we have proposed the presence of superheated pinned vortices at these high temperatures, i.e., vortices which survive at $T>T_c$ only when pinned at $T<T_c$, in which case pinning energy may be large even in the absence of bulk superconductivity.

Unusual as our "vortex" interpretation of the results may be, our observations are remarkably systematic which at least allow us to unambiguously extract a characteristic energy scale, T_s. As discussed above, we believe this is probably associated with superconducting correlations of one form or another but then again it may be associated with some other forms of incipient or actual order. In fact, its close relation to superconductivity, its peak just below and the sudden drop at $x=1/8$ and the abrupt disappearance at $x=0.24$ where it is believed the pseudogap temperature $T^* \approx T_c$, suggests that stripes may be involved in some way.[14,20] The hysteresis could also be associated with some form of orbital ordering which has so far gone undetected. Another possibility, supported by the results in Fig. 8, is the presence of internal superconducting filaments above T_c. Indeed the data may be interpreted using various scenarios.

Certainly more stringent tests are necessary to distinguish the contributions, if more than one, governing the data. If our interpretation is incorrect then Fig. 9, at least, is evidence for a "*magnetic cloud*" hanging over and closely resembling the high-T_c superconducting dome and T_{Nernst} with $T_s=T_c$ when $T^* \approx T_c$. We believe these observations, emerging from our experimental results, are remarkable and deserve further investigation.

In summary, we have observed thermal hysteresis in the normal state magnetization of LSCO, which at least on the basis of the experimental protocols employed in the present study, seems to be related to superconductivity. In the case of $x=0.10$ the hysteresis extends to room temperature. We made an attempt at interpreting our data in terms of superconducting correlations, possibly in the form of pinned vortices, although as discussed before other interpretations, e.g., stripe physics, superconducting filaments may be possible. Also, we cannot rule out the possibility that there is a combination of more than one physical reason causing the observed effects. Either way, our tests indicate the results cannot be due to extrinsic magnetism present only in our samples and that the hysteresis is likely to be linked to superconductivity. They raise new fundamental questions on our understanding of the link between superconductivity and normal state magnetism in high temperature superconductors and the exciting possibility of the presence of superconducting signatures even at room temperature. We hope our work has provided information which will motivate further studies in the search for the presence of superconducting signatures well above T_c and a revised HTS phase diagram linking superconductivity, magnetism, and hole concentrations in a different way and to higher temperatures than presently known.

C.P. thanks A. Castro-Neto, N. Goldenfeld, S.A. Kivelson, J.W. Loram, N.P. Ong, Y. Wang, and T. Xiang for discussions and The Royal Society for financial support. We are indebted to J.R. Cooper and T. Sasagawa for allowing us to perform measurements on their samples to test our results and for helpful communications. M.M. acknowledges the AFRL/PRPS Wright-Patterson Air Force Base, Ohio, for financial support.

*Now at the Departement de Physique de la Matiere Condensee, Universite de Geneve, 24 Quai Ernest-Ansermet, CH-1211 Geneve, Switzerland.

[1] J. G. Bednorz and K. A. Muller, Z. Phys. B: Condens. Matter **64**, 189 (1986).

[2] A. Schilling, M. Cantoni, J. D. Guo, and H. R. Ott, Nature (London) **363**, 56 (1993).

[3] Z. A. Xu, N. P. Ong, Y. Wang, T. Kakeshita, and S. Uchida, Nature (London) **406**, 486 (2000).

[4] Y. Wang, Z. A. Xu, T. Kakeshita, S. Uchida, S. Ono, Y. Ando, and

N. P. Ong, Phys. Rev. B **64**, 224519 (2001).
[5] J. R. Waldram, *Superconductivity of Metals and Cuprates* (Institute of Physics, London, 1996).
[6] N. Kobayashi, K. Hirano, T. Nishizaki, H. Iwasaki, T. Sasaki, S. Awaji, K. Watanabe, H. Asaoka, and H. Takei, Physica C **251**, 255 (1995).
[7] S. Senoussi, J. Phys. III **2**, 1041 (1992).
[8] W. K. Kwok, U. Welp, V. M. Vinokur, S. Fleshler, J. Downey, and G. W. Crabtree, Phys. Rev. Lett. **67**, 390 (1991).
[9] E. W. Collings, *Applied Superconductivity, Metallurgy and Physics of Titanium Alloys* (Plenum, New York, 1985).
[10] I. Iguchi, T. Yamaguchi, and A. Sugimoto, Nature (London) **412**, 420 (2001).
[11] Y. Ando, A. N. Lavrov, and S. Komiya, Phys. Rev. Lett. **90**, 247003 (2003).
[12] J. Zaanen, J. Phys. Chem. Solids **59**, 1769 (1998).
[13] J. M. Tranquada, B. J. Sternlieb, J. D. Axe, Y. Nakamura, and S. Uchida, Nature (London) **375**, 561 (1995).
[14] S. A. Kivelson, E. Fradkin, V. Oganesyan, I. P. Bindloss, J. M. Tranquada, A. Kapitulnik, and C. Howald, Rev. Mod. Phys. **75**, 1201 (2003).
[15] C. Panagopoulos, J. L. Tallon, B. D. Rainford, T. Xiang, J. R. Cooper, and C. A. Scott, Phys. Rev. B **66**, 064501 (2002); Solid State Commun. **126**, 47 (2003).
[16] Y. J. Uemura, G. M. Luke, B. J. Sternlieb, J. H. Brewer, J. F. Carolan, W. N. Hardy, R. Kadono, J. R. Kempton, R. F. Kiefl, S. R. Kreitzman, P. Mulhern, T. M. Riseman, D. L. Williams, B. X. Yang, S. Uchida, H. Takagi, J. Gopalakrishnan, A. W. Sleight, M. A. Subramanian, C. L. Chien, M. Z. Cieplak, G. Xiao, V. Y. Lee, B. W. Statt, C. E. Stronach, W. J. Kossler, and X. H. Yu, Phys. Rev. Lett. **62**, 2317 (1989).
[17] A. Ino, C. Kim, M. Nakamura, T. Yoshida, T. Mizokawa, A. Fujimori, Z.-X. Shen, T. Kakeshita, H. Eisaki, and S. Uchida, Phys. Rev. B **65**, 094504 (2002).
[18] C. Panagopoulos (unpublished).
[19] N. P. Ong and Y. Wang (private communication).
[20] T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999).